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# Analytical Solutions of Temporal Evolution of Populations in Optically-Pumped Atoms with Circularly Polarized Light

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Academic Editor: Young Suh Kim

Received: 10 December 2015; Accepted: 14 March 2016 ; Published: 19 March 2016

**Abstract:** We present an analytical calculation of temporal evolution of populations for optically pumped atoms under the influence of weak, circularly polarized light. The differential equations for the populations of magnetic sublevels in the excited state, derived from rate equations, are expressed in the form of inhomogeneous second-order differential equations with constant coefficients. We present a general method of analytically solving these differential equations, and obtain explicit analytical forms of the populations of the ground state at the lowest order in the saturation parameter. The obtained populations can be used to calculate lineshapes in various laser spectroscopies, considering transit time relaxation.

**Keywords:** second-order differential equations; optical pumping; analytical solutions

**PACS:** 02.30.Hq; 32.80.Xx; 32.30.-r

## 1. Introduction

When an atom is illuminated by single-mode laser light, the populations of the magnetic sublevels and coherences between them exhibit complicated temporal variations. This phenomenon is called optical pumping, which is widely used in the preparation of internal atomic states of interest [1,2]. It has recently been observed that optical pumping affects the lineshapes in saturated absorption spectroscopy (SAS) [3], electromagnetically induced transparency (EIT) [4], and absorption of cold atoms with a  $\Lambda$ -type three-level scheme [5]. Nonlinear effects in optical pumping have also been investigated [6,7].

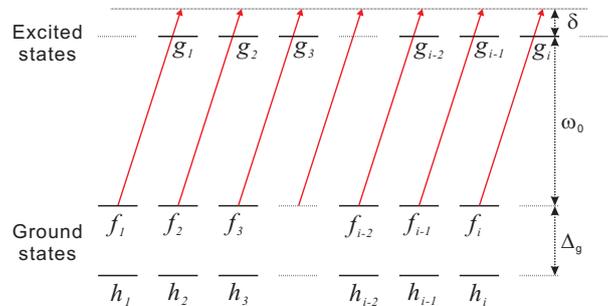
The temporal dynamics of the internal states of an atom are accurately described by density matrix equations [8,9]. In some special cases, however, a simpler method can be employed to solve for the dynamics of the internal states of the atom, using rate equations [10,11]. Furthermore, when the intensity of light is weak, the rate equations can be solved analytically [12–15]. These analytical solutions are practically very useful; once they are obtained, it is readily possible to obtain analytically computed quantities such as the absorption coefficient of a probe beam and lineshape functions in nonlinear laser spectroscopy. We have previously reported analytical solutions for SAS [16,17] and polarization spectroscopy (PS) [18].

Interestingly, the equations governing the temporal dynamics of populations at the weak intensity limit are homogeneous or inhomogeneous second-order linear differential equations (DEs) with constant coefficients [12–15]. Unlike the harmonic oscillator in mechanics, where under- or over-damped motions are observed [19], the equations for optical pumping show only over-damped behaviors. However, this system exhibits a variety of inhomogeneous DEs. In a recent publication, we reported the method of solving these equations analytically, in the context of

a pedagogical description of the method of solving inhomogeneous DEs [15]. Although the method is straightforward in principle, it is not easy to obtain analytical solutions for complicated atomic structures, such as Cs. Extending the previous study [15], in this paper, we present a general method of analytically solving the DEs for such a complicated atom.

## 2. Theory

The energy level diagram under consideration is shown in Figure 1. Since alkali-metal atoms are considered, there are two ground states with  $F_g = I + 1/2$  and  $F_g = I - 1/2$  ( $I$ : nuclear spin angular momentum quantum number). We consider a  $\sigma^+$  polarized weak laser beam, whose Rabi frequency is  $\Omega$  and optical frequency is  $\omega = \omega_0 + \delta$  ( $\omega_0$  is the resonance frequency and  $\delta$  is the laser frequency detuning). We assume that the laser frequency is tuned to the transition from one of the two ground states (in Figure 1, the state  $F_g = I + 1/2$ ). Then, the other ground state (in Figure 1, the state  $F_g = I - 1/2$ ) is not excited by laser light, and can be populated by spontaneous emission from the excited state when the optical transition is not cycling. The populations (and the states themselves) of the magnetic sublevels in the excited, upper ground, and lower ground states are labeled, respectively, as  $g_i$ ,  $f_i$ , and  $h_i$  with  $i = 1, 2, \dots$ .



**Figure 1.** An energy level diagram for an optically pumped atom under the influence of circularly polarized light.

The internal dynamics of the atom can be described by the density matrix equation in the frame rotating with frequency  $\omega$ :

$$\dot{\rho} = - (i/\hbar) [H, \rho] + \dot{\rho}_{\text{sp}}, \quad (1)$$

where  $\rho$  is the density operator. In Equation (1), the Hamiltonian,  $H$ , is given by

$$H = - \sum_j \hbar \delta |g_j\rangle \langle g_j| - \sum_j \hbar \Delta_g |h_j\rangle \langle h_j| - \frac{\hbar \Omega}{2} \sum_j C_j^j |g_j\rangle \langle f_j| + \text{h.c.}, \quad (2)$$

where  $\Delta_g$  is the hyperfine splitting between the two ground states and h.c. denotes the harmonic conjugate. In Equation (2), the first two terms in the right-hand side represent the bare atomic Hamiltonian and the rest terms denote the atom-photon interaction Hamiltonian [20].  $C_i^j$  is the normalized transition strength between the states  $f_i$  and  $g_j$ , and  $R_i^j \equiv (C_i^j)^2$  is given below (Equation (13)). In Equation (1),  $\dot{\rho}_{\text{sp}}$  represents spontaneous emission term, whose matrix representations are given by:

$$\begin{aligned} \langle g_i | \dot{\rho}_{\text{sp}} | g_j \rangle &= -\Gamma \langle g_i | \rho | g_j \rangle, \\ \langle g_i | \dot{\rho}_{\text{sp}} | f_j \rangle &= -\frac{\Gamma}{2} \langle g_i | \rho | f_j \rangle, \quad \langle g_i | \dot{\rho}_{\text{sp}} | h_j \rangle = -\frac{\Gamma}{2} \langle g_i | \rho | h_j \rangle, \\ \langle f_i | \dot{\rho}_{\text{sp}} | f_j \rangle &= \Gamma \sum_{\epsilon=-2}^0 C_i^{i+\epsilon} C_j^{j+\epsilon} \langle g_{i+\epsilon} | \rho | g_{j+\epsilon} \rangle, \\ \langle h_i | \dot{\rho}_{\text{sp}} | h_j \rangle &= \Gamma \sum_{\epsilon=-2}^0 D_i^{i+\epsilon} D_j^{j+\epsilon} \langle g_{i+\epsilon} | \rho | g_{j+\epsilon} \rangle, \end{aligned} \quad (3)$$

and  $\langle \mu | \dot{\rho}_{\text{sp}} | \nu \rangle = \langle \nu | \dot{\rho}_{\text{sp}} | \mu \rangle^*$  when  $\mu \neq \nu$ , where  $\Gamma$  is the decay rate of the excited state.  $D_i^j$  is the normalized transition strength between the states  $h_i$  and  $g_j$ , and  $T_i^j \equiv (D_i^j)^2$  is also given below (Equation (13)). Inserting Equations (2) and (3) into Equation (1), we can obtain the following differential equations for the optical coherences and populations:

$$\langle g_i | \dot{\rho} | f_i \rangle = \left( i\delta - \frac{\Gamma}{2} \right) \langle g_i | \rho | f_i \rangle + \frac{i}{2} C_i^i \Omega (g_i - f_i), \quad (4)$$

$$\dot{g}_i = -\Gamma g_i + \frac{i}{2} C_i^i \Omega (\langle g_i | \rho | f_i \rangle - \langle f_i | \rho | g_i \rangle), \quad (5)$$

$$\dot{f}_i = \Gamma \sum_{j=i-2}^i (C_i^j)^2 g_j - \frac{i}{2} C_i^i \Omega (\langle g_i | \rho | f_i \rangle - \langle f_i | \rho | g_i \rangle), \quad (6)$$

$$\dot{h}_i = \Gamma \sum_{j=i-2}^i (D_i^j)^2 g_j, \quad (7)$$

where we use simplified expressions for the populations:  $\langle g_i | \rho | g_i \rangle = g_i$ ,  $\langle f_i | \rho | f_i \rangle = f_i$ , and  $\langle h_i | \rho | h_i \rangle = h_i$ . In Equations (4)–(7), we assume that  $\langle g_i | \rho | h_i \rangle = 0$  because  $\Delta_g$  is much larger than  $|\delta|$  and  $\Gamma$ . We note that, because the polarization of light is  $\sigma^+$ , and therefore the Zeeman coherences between the magnetic sublevels in the excited and ground states disappear.

In Equation (4), the characteristic decay rate of the optical coherence is  $\Gamma/2$ , which is much larger than the characteristic decay rate of the populations ( $\sim s\Gamma$ ; see Equation (12) below for definition of  $s$ ). Thus, the optical coherences evolve much faster than the populations, which is called the rate equation approximation [21]. Owing to this rate equation approximation,  $\langle g_i | \rho | f_i \rangle$  can be expressed in terms of the populations as follows by letting  $\langle g_i | \dot{\rho} | f_i \rangle = 0$ :

$$\langle g_i | \rho | f_i \rangle = \frac{C_i^i \Omega}{i\Gamma + 2\delta} (f_i - g_i). \quad (8)$$

Then, inserting Equation (8) and its complex conjugate into Equations (5)–(7), we can obtain the following rate equations for the populations:

$$\dot{f}_i = -\frac{\Gamma}{2} s R_i^i (f_i - g_i) + \sum_{j=i-2}^i \Gamma R_i^j g_j, \quad (9)$$

$$\dot{g}_i = \frac{\Gamma}{2} s R_i^i (f_i - g_i) - \Gamma g_i, \quad (10)$$

$$\dot{h}_i = \sum_{j=i-2}^i \Gamma T_i^j g_j, \quad (11)$$

for  $i = 1, 2, \dots$ . In Equations (9)–(11),  $s$  is the saturation parameter, which is given by

$$s = \frac{\Omega^2/2}{\delta^2 + \Gamma^2/4}, \quad (12)$$

and  $R_i^j = (C_i^j)^2$  and  $T_i^j = (D_i^j)^2$ . We note that  $s$  is a function of both the  $\delta$  and Rabi frequency. Notably, the reference of the frequency detuning differs, depending on the transition line considered.

When  $i$  and  $j$  refer to the states  $|F_g, m_g\rangle$  and  $|F_e, m_e\rangle$ , respectively, the transition strength ( $R_i^j$ ) is given by

$$R_{F_g, m_g}^{F_e, m_e} = (2L_e + 1)(2J_e + 1)(2J_g + 1)(2F_e + 1)(2F_g + 1) \times \left[ \left\{ \begin{matrix} L_e & J_e & S \\ J_g & L_g & 1 \end{matrix} \right\} \left\{ \begin{matrix} J_e & F_e & I \\ F_g & J_g & 1 \end{matrix} \right\} \left( \begin{matrix} F_g & 1 & F_e \\ m_g & m_e - m_g & -m_e \end{matrix} \right) \right]^2, \quad (13)$$

where  $L$  and  $S$  denote the orbital and electron spin angular momenta, respectively, and the curly (round) brackets represent the  $6J$  ( $3J$ ) symbol.  $T_i^j$  are similarly obtained by using different  $F_g$  values in Equation (13).

The explicit form of Equation (9) is given by

$$\dot{f}_i = \frac{\Gamma}{2} s R_i^i (g_i - f_i) + \Gamma \left( R_i^{i-2} g_{i-2} + R_i^{i-1} g_{i-1} + R_i^i g_i \right), \quad (14)$$

and  $f_i$  can be expressed in terms of  $\dot{g}_i$  and  $g_i$  from Equation (10) at the lowest order in  $s$  as follows:

$$f_i = \frac{2}{\Gamma s R_i^i} (\dot{g}_i + \Gamma g_i). \quad (15)$$

Insertion of Equations (10) and (15) into Equation (14) yields the following DE for  $g_i$ :

$$\begin{aligned} \ddot{g}_i + \Gamma \left( 1 + \frac{s}{2} R_i^i \right) \dot{g}_i + \frac{s}{2} \Gamma^2 R_i^i (1 - R_i^i) g_i \\ = \frac{s}{2} \Gamma^2 R_i^{i-2} R_i^i g_{i-2} + \frac{s}{2} \Gamma^2 R_i^{i-1} R_i^i g_{i-1}. \end{aligned} \quad (16)$$

when  $i = 1$ , the right-hand side of Equation (16) vanishes. Therefore, Equation (16) becomes a homogeneous DE. In contrast, when  $i \neq 1$ , Equation (16) becomes an inhomogeneous DE because the right-hand side terms are functions of  $g_i$ .

We solve Equation (16) from  $i = 1$  consecutively. As is well-known, the solution of Equation (16) consists of two parts: a homogeneous solution and a particular solution. We first find the solutions of the homogeneous equation by inserting the equation  $g_i \sim e^{\lambda \Gamma t}$  into Equation (16). Then, we have two values ( $\lambda_{2i-1}$ ,  $\lambda_{2i}$ ) for  $\lambda$  as follows:

$$\lambda_{2i-1(2i)} = \frac{1}{4} \left( -2 - s R_i^i - (+) \sqrt{4 - 4s R_i^i + s(8 + s) (R_i^i)^2} \right),$$

which can be approximated as follows in the weak intensity limit:

$$\lambda_{2i-1} \simeq -1 - \frac{s}{2} (R_i^i)^2, \quad \lambda_{2i} \simeq -\frac{s}{2} R_i^i (1 - R_i^i).$$

We consider the case of  $i = 1$  in Equation (16). Then, the solution is given by

$$g_1 = C_{1,1} e^{\lambda_1 \Gamma t} + C_{1,2} e^{\lambda_2 \Gamma t},$$

where the coefficients  $C_{1,1}$  and  $C_{1,2}$  should be determined using the initial conditions. In the case of  $i = 2$ , the right-hand side in Equation (16) contains the terms of  $e^{\lambda_1 \Gamma t}$  and  $e^{\lambda_2 \Gamma t}$ . Therefore,  $g_2$  has four exponential terms:

$$g_2 = C_{2,1} e^{\lambda_1 \Gamma t} + C_{2,2} e^{\lambda_2 \Gamma t} + C_{2,3} e^{\lambda_3 \Gamma t} + C_{2,4} e^{\lambda_4 \Gamma t},$$

where the coefficients should also be determined. Therefore, we can express  $g_j$  generally as follows:

$$g_j = \sum_{k=1}^{2j} C_{j,k} e^{\lambda_k \Gamma t}. \tag{17}$$

We find  $C_{j,k}$  with  $k = 1, 2, \dots, 2j$  by means of recursion relations; *i.e.*,  $C_{j,k}$  are expressed in terms of  $C_{i,l}$  with  $i < j$  and  $l = 1, 2, \dots, 2i$ . Inserting Equation (17) into Equation (16), we obtain

$$\begin{aligned} g_i &= C_{i,2i-1} e^{\lambda_{2i-1} \Gamma t} + C_{i,2i} e^{\lambda_{2i} \Gamma t} \\ &+ \sum_{k=1}^{2(i-1)} \frac{(s/2) R_i^{i-1} R_i^i C_{i-1,k}}{\lambda_k^2 + \lambda_k + \frac{s}{2} R_i^i (1 + \lambda_k - R_i^i)} e^{\lambda_k \Gamma t} \\ &+ \sum_{k=1}^{2(i-2)} \frac{(s/2) R_i^{i-2} R_i^i C_{i-2,k}}{\lambda_k^2 + \lambda_k + \frac{s}{2} R_i^i (1 + \lambda_k - R_i^i)} e^{\lambda_k \Gamma t}. \end{aligned} \tag{18}$$

Comparing Equations (17) and (18) gives

$$C_{i,k} = \frac{(s/2) R_i^i (R_i^{i-1} C_{i-1,k} + R_i^{i-2} C_{i-2,k})}{\lambda_k^2 + \lambda_k + \frac{s}{2} R_i^i (1 + \lambda_k - R_i^i)}, \tag{19}$$

for  $k = 1, 2, \dots, 2(i-2)$ ,

$$C_{i,k} = \frac{(s/2) R_i^{i-1} R_i^i C_{i-1,k}}{\lambda_k^2 + \lambda_k + \frac{s}{2} R_i^i (1 + \lambda_k - R_i^i)}, \tag{20}$$

for  $k = 2i-3$  and  $2(i-1)$ .

The remaining two coefficients,  $C_{i,2i-1}$  and  $C_{i,2i}$ , can be derived from Equation (18) using two initial conditions for  $g_i(0)$  and  $\dot{g}_i(0)$ :

$$g_i(0) = 0, \quad \dot{g}_i(0) = \frac{s}{2} p_0 R_i^i,$$

where  $p_0$  is the population of each sublevel in the ground state at equilibrium, which is equal to  $1/[2(2I + 1)]$ . Then, the results are given by

$$C_{i,2i-1} = \frac{1}{2Q_i} [2(A_i + 2A'_i + B_i + 2B'_i) + (A_i + B_i - 2p_0) s R_i^i] - \frac{A_i + B_i}{2}, \tag{21}$$

$$C_{i,2i} = -\frac{1}{2Q_i} [2(A_i + 2A'_i + B_i + 2B'_i) + (A_i + B_i - 2p_0) s R_i^i] - \frac{A_i + B_i}{2}, \tag{22}$$

where

$$\begin{aligned} Q_i &= \sqrt{4 + s R_i^i (-4 + (8 + s) R_i^i)}, \\ A_i &= \sum_{k=1}^{2(i-1)} \frac{(s/2) R_i^{i-1} R_i^i C_{i-1,k}}{\lambda_k^2 + \lambda_k + \frac{s}{2} R_i^i (1 + \lambda_k - R_i^i)}, \text{ for } i \geq 2 \\ B_i &= \sum_{k=1}^{2(i-2)} \frac{(s/2) R_i^{i-2} R_i^i C_{i-2,k}}{\lambda_k^2 + \lambda_k + \frac{s}{2} R_i^i (1 + \lambda_k - R_i^i)}, \text{ for } i \geq 3, \\ A'_i &= \sum_{k=1}^{2(i-1)} \frac{(s/2) R_i^{i-1} R_i^i \lambda_k C_{i-1,k}}{\lambda_k^2 + \lambda_k + \frac{s}{2} R_i^i (1 + \lambda_k - R_i^i)}, \text{ for } i \geq 2 \\ B'_i &= \sum_{k=1}^{2(i-2)} \frac{(s/2) R_i^{i-2} R_i^i \lambda_k C_{i-2,k}}{\lambda_k^2 + \lambda_k + \frac{s}{2} R_i^i (1 + \lambda_k - R_i^i)}, \text{ for } i \geq 3, \end{aligned}$$

and

$$A_1 = 0, \quad A'_1 = 0, \quad B_1 = B_2 = 0, \quad \text{and} \quad B'_1 = B'_2 = 0.$$

The coefficients in  $g_i$  from  $g_1$  can be obtained by successively using the recursion relations in Equations (19)–(22). Once  $g_i$  are obtained,  $f_i$  can be obtained using Equation (15). Up to the lowest order in  $s$ , the result is given by

$$f_i = \sum_{k=1}^i \frac{2C_{i,2k}}{sR_i^i} e^{\lambda_{2k}\Gamma t}. \quad (23)$$

Since  $\lambda_k \sim -1$  for odd  $k$ ,  $g_i$  can be expressed as follows:

$$g_i = \sum_{k=1}^i \left( C_{i,2k-1} e^{-\Gamma t} + C_{i,2k} e^{\lambda_{2k}\Gamma t} \right). \quad (24)$$

Taking the derivative of Equation (24) with respect to time and letting  $t = 0$ , we have

$$\dot{g}_i(0) = - \sum_{k=1}^i C_{i,2k-1},$$

up to the first order in  $s$ , since  $\lambda_{2k}$  ( $k = 1, 2, \dots, i$ ) are already in the first order in  $s$ . Because one of the initial conditions is  $\dot{g}_i(0) = sp_0 R_i^i / 2$ , and  $g_i(0) = \sum_{k=1}^i (C_{i,2k-1} + C_{i,2k}) = 0$  from the other initial condition, we obtain the following equations:

$$\sum_{k=1}^i C_{i,2k-1} = - \sum_{k=1}^i C_{i,2k} = -\frac{s}{2} p_0 R_i^i. \quad (25)$$

Using the relations in Equations (23) and (25), we find the simplified form of  $g_i$  as follows:

$$g_i = \frac{R_i^i s}{2} \left( f_i - p_0 e^{-\Gamma t} \right). \quad (26)$$

We obtain the populations of the sublevels in the ground state, which are not excited by laser light. The one or two magnetic sublevels with higher magnetic quantum numbers correspond to this case. We can easily obtain analytical populations by integrating the populations spontaneously transferred from the excited state, and the result is given by

$$f_i = p_0 + \sum_{k=1}^{i-2} R_i^{i-2} C_{i-2,2k} \frac{e^{\lambda_{2k}\Gamma t} - 1}{\lambda_{2k}} + \sum_{k=1}^{i-1} R_i^{i-1} C_{i-1,2k} \frac{e^{\lambda_{2k}\Gamma t} - 1}{\lambda_{2k}}. \quad (27)$$

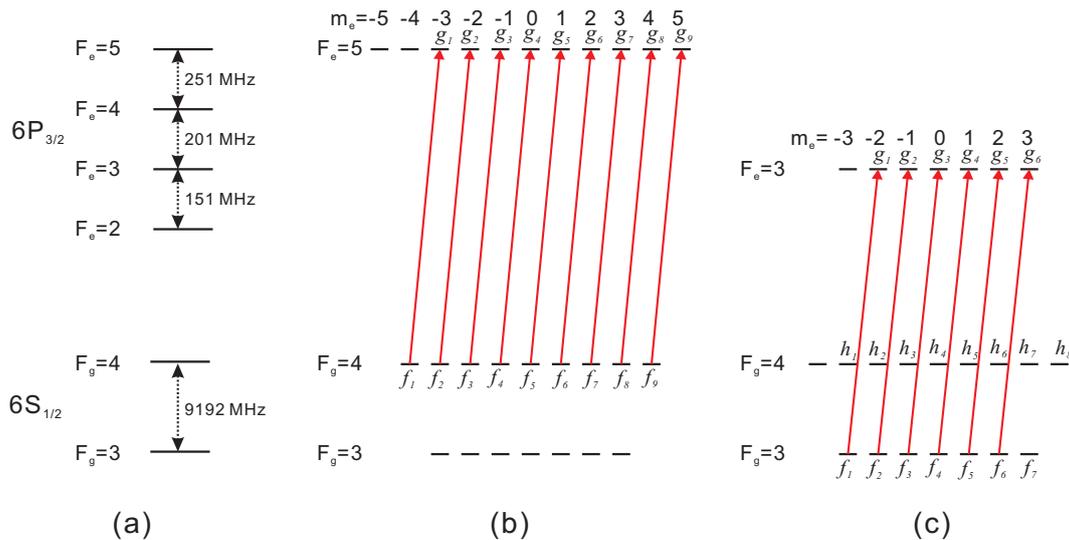
In several cases of atomic transition systems,  $\lambda_k$  can duplicate, and the method of solving particular solutions given in Equation (18) no longer holds. We may solve for the particular solutions using the method presented in our previous paper [15]. However, it is also possible to solve by intentionally modifying  $\lambda_k$  to satisfy the conditions that all  $\lambda_k$  are unique. One possible method is setting  $R_i^j \rightarrow R_i^j + \delta_{i,j} j \epsilon$ , where  $\epsilon$  is a constant that is taken as zero at the final stage of the calculation. Although this method is not novel, it is very efficient.

The populations ( $h_i$ ) of the sublevels in the ground state, which are not excited by laser light, can be easily obtained analytically by integrating the populations spontaneously transferred from the excited state (Equation (11)), and the result is given by

$$h_i = p_0 + \sum_{l=-2}^0 \sum_{k=1}^{i+l} T_i^{i+l} C_{i+l,2k} \frac{e^{\lambda_{2k}\Gamma t} - 1}{\lambda_{2k}}. \quad (28)$$

### 3. Calculated Results

Based on the method developed in Section 2, here we present the calculated results of the populations for the two transition schemes: (i)  $F_g = 4 \rightarrow F_e = 5$  and (ii)  $F_g = 3 \rightarrow F_e = 3$  for the D2 line of Cs. The energy level diagram for the Cs-D2 line is shown in Figure 2a, and the energy level diagrams for these two transitions are shown in Figure 2b,c. Owing to the large hyperfine splitting in the excited states, it is justifiable to neglect the off-resonant transitions; *i.e.*, the  $F_g = 4 \rightarrow F_e = 4$  and  $F_g = 4 \rightarrow F_e = 3$  transitions can be neglected when the laser light is tuned to the  $F_g = 4 \rightarrow F_e = 5$  transition line. Although it is in principle possible to include the off-resonant transitions in the analytical calculation of the populations [13], the complicated analytical solutions may not be practically useful.



**Figure 2.** (a) Energy level diagram of the Cs-D2 line. (b) Energy level diagrams for the  $F_g = 4 \rightarrow F_e = 5$  cycling transition line and (c) for the  $F_g = 3 \rightarrow F_e = 3$  transition line illuminated by  $\sigma^+$  polarized laser light.

#### 3.1. Results for the $F_g = 4 \rightarrow F_e = 5$ Transition

The  $F_g = 4 \rightarrow F_e = 5$  transition shown in Figure 2b is cycling, and is used in many experiments, such as laser cooling and trapping [22]. Because  $\sigma^+$  polarized laser light is illuminated, the sublevels with  $m_e = -5$  and  $-4$  are not optically excited. The normalized transition strengths, for the transitions presented in Figure 2b, are given by

$$\begin{aligned} & (R_1^1, R_2^2, R_3^3, R_4^4, R_5^5, R_6^6, R_7^7, R_8^8, R_9^9) \\ &= \left( \frac{1}{45}, \frac{1}{15}, \frac{2}{15}, \frac{2}{9}, \frac{1}{3}, \frac{7}{15}, \frac{28}{45}, \frac{4}{5}, 1 \right). \end{aligned}$$

For the transition for  $i = 1$ , we obtain  $\lambda_1 \simeq -1$  and  $\lambda_2 \simeq -\frac{22}{2025}s$ , and

$$C_{1,1} = -\frac{s}{1440}, \quad C_{1,2} = \frac{s}{1440}.$$

Thus, using Equation (23), we obtain

$$f_1 = \frac{1}{16} e^{-22s\Gamma t/2025}.$$

The  $\lambda_4$  for the transition for  $i = 2$  is approximately given by  $-\frac{7}{225}s$ , and the coefficients are given by

$$C_{2,1} = \frac{s}{240}, \quad C_{2,2} = \frac{s}{2460},$$

$$C_{2,3} = -\frac{s}{160}, \quad C_{2,4} = \frac{11}{6560}s.$$

Therefore, we have

$$f_2 = \frac{1}{82}e^{-22s\Gamma t/2025} + \frac{33}{656}e^{-7s\Gamma t/225}.$$

The remaining  $\lambda_{2k}$  ( $k = 2, \dots, 9$ ) values are given by

$$(\lambda_6, \lambda_8, \lambda_{10}, \lambda_{12}, \lambda_{14}, \lambda_{16}, \lambda_{18})$$

$$= \left( -\frac{13}{225}s, -\frac{7}{81}s, -\frac{s}{9}, -\frac{28}{225}s, -\frac{238}{2025}s, -\frac{2}{25}s, 0 \right),$$

and the remaining populations are explicitly given by

$$f_3 = \frac{413}{31160}e^{-22\tau/2025} + \frac{77}{2624}e^{-7\tau/225} + \frac{121}{6080}e^{-13\tau/225},$$

$$f_4 = \frac{2317}{264860}e^{-22\tau/2025} + \frac{693}{20992}e^{-7\tau/225} + \frac{1089}{44080}e^{-13\tau/225} - \frac{1001}{252416}e^{-7\tau/81},$$

$$f_5 = \frac{25577}{3072376}e^{-22\tau/2025} + \frac{4235}{125952}e^{-7\tau/225} + \frac{5203}{141056}e^{-13\tau/225}$$

$$- \frac{5005}{504832}e^{-7\tau/81} - \frac{143}{22272}e^{-\tau/9},$$

$$f_6 = \frac{148693}{17666162}e^{-22\tau/2025} + \frac{1925}{47232}e^{-7\tau/225} + \frac{2057}{35264}e^{-13\tau/225}$$

$$- \frac{1625}{63104}e^{-7\tau/81} - \frac{715}{16704}e^{-\tau/9} + \frac{13}{552}e^{-28\tau/225},$$

$$f_7 = \frac{921751}{89260608}e^{-22\tau/2025} + \frac{2519}{41984}e^{-7\tau/225} + \frac{891}{7424}e^{-13\tau/225}$$

$$- \frac{49075}{504832}e^{-7\tau/81} - \frac{5555}{7424}e^{-\tau/9} - \frac{273}{736}e^{-28\tau/225} + \frac{209}{192}e^{-238\tau/2025},$$

$$f_8 = \frac{39041249}{2119939440}e^{-22\tau/2025} + \frac{1561}{10496}e^{-7\tau/225} + \frac{225071}{352640}e^{-13\tau/225} + \frac{219275}{126208}e^{-7\tau/81}$$

$$+ \frac{9955}{3712}e^{-\tau/9} + \frac{3367}{3680}e^{-28\tau/225} - \frac{77}{24}e^{-238\tau/2025} - \frac{459}{160}e^{-2\tau/25},$$

$$f_9 = \frac{9}{16} - \frac{1205666281}{8479757760}e^{-22\tau/2025} - \frac{74771}{188928}e^{-7\tau/225} - \frac{316701}{352640}e^{-13\tau/225}$$

$$- \frac{404009}{252416}e^{-7\tau/81} - \frac{62953}{33408}e^{-\tau/9} - \frac{3133}{5520}e^{-28\tau/225} + \frac{407}{192}e^{-238\tau/2025} + \frac{459}{160}e^{-2\tau/25},$$

where we use a simplified notation:  $\tau \equiv s\Gamma t$ . Since the  $F_g = 4 \rightarrow F_e = 5$  transition is cycling, the populations in the magnetic sublevels in the  $F_g = 3$  ground state remain at their equilibrium value,  $1/16$ . It should be also noted that the sum of the ground state populations is conserved, *i.e.*,

$$\sum_{i=1}^9 f_i = \frac{9}{16}.$$

From Equation (26), the populations of the sublevels in the excited state can be expressed in terms of the populations in the ground state as follows:

$$g_i = \frac{R_i^i s}{2} \left( f_i - \frac{1}{16} e^{-\Gamma t} \right).$$

The constants in  $f_9$  and  $g_9$  can be accurately calculated using Equation (10). In the steady-state regime, all the populations except  $f_9$  and  $g_9$  vanish, and these satisfy the following equations:

$$\frac{\Gamma}{2} s [f_9(\infty) - g_9(\infty)] - \Gamma g_9(\infty) = 0, \quad f_9(\infty) + g_9(\infty) = \frac{9}{16},$$

with  $R_9^9 = 1$ . Then, we have

$$f_9(\infty) = \frac{9(2+s)}{32(1+s)}, \quad g_9(\infty) = \frac{9s}{32(1+s)},$$

which can be used in a more accurate calculation of the SAS spectrum.

### 3.2. Results for the $F_g = 3 \rightarrow F_e = 3$ Transition

Now we present the calculated results of the populations for the  $F_g = 3 \rightarrow F_e = 3$  transition of the D2 line of Cs. The energy level diagram for the transition is shown in Figure 2c. The sublevel of the excited state with  $m_e = -3$  is not optically excited, and thus the sublevel of the upper-ground state with  $m_g = -4$  is not filled by spontaneous emission. We also obtain the solutions for the populations in the other ground state ( $F_g = 4$ ). To prevent the duplication of the transition strengths in this transition, we introduce  $\epsilon$  so that the transition strengths are given explicitly by

$$\begin{aligned} & (R_1^1, R_2^2, R_3^3, R_4^4, R_5^5, R_6^6) \\ &= \left( \frac{3}{16} + \epsilon, \frac{5}{16} + 2\epsilon, \frac{3}{8} + 3\epsilon, \frac{3}{8} + 4\epsilon, \frac{5}{16} + 5\epsilon, \frac{3}{16} + 6\epsilon \right). \end{aligned}$$

We take  $\epsilon \rightarrow 0$  at the final stage of the calculation. The  $\lambda_{2k}$  ( $k = 1, \dots, 6$ ) values at  $\epsilon \rightarrow 0$  are given by

$$\begin{aligned} & (\lambda_2, \lambda_4, \lambda_6, \lambda_8, \lambda_{10}, \lambda_{12}) \\ &= \left( -\frac{39}{512}s, -\frac{55}{512}s, -\frac{15}{128}s, -\frac{15}{128}s, -\frac{55}{512}s, -\frac{39}{512}s \right). \end{aligned}$$

We first find various  $C_{ik}$  values using the recursion relations in Equations (19)–(22). For the transition for  $i = 1$ , we obtain

$$C_{1,1} = -\frac{3}{512}s, \quad C_{1,2} = \frac{3}{512}s;$$

thus, using Equation (23), we obtain

$$f_1 = \frac{1}{16} e^{-39s\Gamma t/512}.$$

Using a similar method, we can obtain  $f_2$  and  $f_3$  as follows:

$$\begin{aligned} f_2 &= \frac{3}{64} e^{-39\tau/512} + \frac{1}{64} e^{-55\tau/512}, \\ f_3 &= \frac{25}{448} e^{-39\tau/512} + \frac{1}{64} e^{-55\tau/512} - \frac{1}{112} e^{-15\tau/128}, \end{aligned}$$

where the simplified notation,  $\tau \equiv s\Gamma t$ , is used. In the calculation of  $f_4$ , because  $\lambda_6$  and  $\lambda_8$  are equal,  $f_4$  may contain the term  $\sim \tau e^{-15\tau/128}$ . However, because the transition between  $g_3$  and  $f_4$  is prohibited, the particular solution for  $f_4$  does not contain the term  $\sim \tau e^{-15\tau/128}$ . In contrast,  $f_5$ ,  $f_6$ , and  $f_7$  contain the terms proportional to  $\tau$ . The results for  $f_4$ ,  $f_5$ , and  $f_6$  are explicitly given by

$$\begin{aligned} f_4 &= \frac{15}{224}e^{-39\tau/512} + \frac{3}{32}e^{-55\tau/512} - \frac{11}{112}e^{-15\tau/128}, \\ f_5 &= \frac{135}{896}e^{-39\tau/512} + \left(-\frac{173}{640} + \frac{9\tau}{4096}\right)e^{-55\tau/512} + \frac{51}{280}e^{-15\tau/128}, \\ f_6 &= \left(\frac{269}{12544} + \frac{1125\tau}{114688}\right)e^{-39\tau/512} \\ &+ \left(\frac{19}{256} - \frac{45\tau}{16384}\right)e^{-55\tau/512} - \frac{13}{392}e^{-15\tau/128}. \end{aligned}$$

Since  $f_7$  is not excited by laser light, using Equation (27) yields,

$$\begin{aligned} f_7 &= \frac{68971}{327184} - \left(\frac{343323}{2119936} + \frac{10125\tau}{1490944}\right)e^{-39\tau/512} \\ &+ \left(\frac{1371}{30976} + \frac{135\tau}{180224}\right)e^{-55\tau/512} - \frac{3}{98}e^{-15\tau/128}. \end{aligned}$$

The populations of the sublevels in the excited state, using Equation (26), can be expressed as follows:

$$g_i = \frac{R_i^i s}{2} \left( f_i - \frac{1}{16}e^{-\Gamma t} \right).$$

The populations of the sublevels in the ground state  $F_g = 4$  can be obtained using Equation (28), and are presented in the appendix.

#### 4. Conclusions

We have presented a general method of solving homogeneous or inhomogeneous second-order DEs corresponding to the optical pumping phenomenon with  $\sigma^+$  polarized laser light. Unlike the harmonic oscillator in mechanics or electrical circuits, this system only exhibits over-damped behavior. Although the method of solving inhomogeneous DEs with constant coefficients is straightforward in principle, obtaining accurate analytical solutions for the equations related to optically pumped atoms, in particular, those with complicated atomic structures, such as Cs, is cumbersome. Our method of solving the DEs provides an easy way to obtain analytical solutions at the weak intensity limit. This method is general and applicable to most atoms. As stated in Section 1, the obtained analytical form of the populations can be used in the calculation of spectroscopic lineshapes such as in saturated absorption spectroscopy (SAS) [16,17] and polarization spectroscopy (PS) [18]. Calculations of SAS and PS for Cs atoms are in progress.

**Acknowledgments:** This research was supported by Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Science, ICT and future Planning (2014R1A2A2A01006654).

**Conflicts of Interest:** The authors declare no conflict of interest.

#### Appendix

When the laser frequency is tuned to the  $F_g = 3 \rightarrow F_e = 3$  transition (Figure 2c), the populations of the sublevels in the ground state  $F_g = 4$  are given by

$$\begin{aligned}
h_1 &= \frac{23}{312} - \frac{7}{624} e^{-39\tau/512}, \\
h_2 &= \frac{93}{1144} - \frac{41}{2496} e^{-39\tau/512} - \frac{5}{2112} e^{-55\tau/512}, \\
h_3 &= \frac{895}{10296} - \frac{1109}{52416} e^{-39\tau/512} - \frac{3}{704} e^{-55\tau/512} + \frac{1}{1008} e^{-15\tau/128}, \\
h_4 &= \frac{235}{2574} - \frac{685}{26208} e^{-39\tau/512} - \frac{19}{1760} e^{-55\tau/512} + \frac{41}{5040} e^{-15\tau/128}, \\
h_5 &= \frac{10727}{113256} - \frac{3475}{104832} e^{-39\tau/512} \\
&\quad - \left( \frac{2641}{232320} + \frac{3\tau}{45056} \right) e^{-55\tau/512} + \frac{31}{2520} e^{-15\tau/128}, \\
h_6 &= \frac{143477}{1472328} - \left( \frac{843497}{19079424} + \frac{125\tau}{1490944} \right) e^{-39\tau/512} \\
&\quad + \left( \frac{401}{30976} - \frac{45\tau}{180224} \right) e^{-55\tau/512} - \frac{13}{3528} e^{-15\tau/128}, \\
h_7 &= \frac{293731}{2944656} - \left( \frac{147347}{2725632} + \frac{125\tau}{212992} \right) e^{-39\tau/512} \\
&\quad + \left( \frac{7889}{154880} - \frac{63\tau}{180224} \right) e^{-55\tau/512} - \frac{43}{1260} e^{-15\tau/128}, \\
h_8 &= \frac{299023}{2944656} - \left( \frac{24497}{681408} + \frac{125\tau}{53248} \right) e^{-39\tau/512} \\
&\quad + \left( -\frac{959}{116160} + \frac{21\tau}{45056} \right) e^{-55\tau/512} + \frac{13}{2520} e^{-15\tau/128}.
\end{aligned}$$

Finally, we note that the sum of the populations is conserved, *i.e.*,

$$\frac{1}{16} + \sum_{i=1}^7 f_i + \sum_{i=1}^8 h_i = 1,$$

where 1/16 is the population at the sublevel  $m_g = -4$  in the upper ground state.

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